Scientific Report

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INTELBIOMAT stay at Zagreb

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The aim of the research stay was to initiate a joint European effort for novek thermoelectrics, including scientists from different European Countries. Through the research stay in Zagreb the concept for such a cooperative effort has been laid. As the most promising materials class for thermoelectric devices clathrates have been identified which, for industrial relevant applications, should be based on Si. In the following we will outline the concept.

Background

Thermoelectric devices are heat engines that convert thermal energy into electricity and vice versa. This allows a direct harvesting of the waste heat, coverting it into electricity. The efficiency of such processes, however, is material dependent and most currently available thermoelectrics are ineficient at low temperatures and instable at high temperatures. The search for new efficient thermoelectrics is trying to exploit the concept of Phonon Liquid and Electron Crystal (PLEC) which tries to combine a good electric conductivity with poor thermal conductivity. Typical examples are provided by compounds with a rigid structure, responsible for good electrical conductivity but with heavy atoms filling large cages, such that rattling modes give rise to phonon scattering that decreases the lattice part of the thermal conductivity to values as low as 2 W/m.K. Dresselhaus et al. explored the phase space of lower dimensionalities demonstrating that the figure of merit ZT might reach values of about 2 in multilayer thin films.

Clathrates as thermoelectrics

Intermetallic clathrates are promising candidates for the PLEC concept exhibiting low values of thermal conductivity and large Seebeck coefficients due to reduced carrier concentrations. Clathrates form a rich class of cage-forming compounds, where tailoring of physical properties due to substitution or doping may create favourable electronic conditions to reach a competitive thermoelectric performance.

Nanostructuring for phonon engineering

Nanostructuring has shown to be a successful concept to further improve thermoelectric performance primarily by introducing various appropriate length scales causing a dramatic drop of thermal conductivity and, moreover, quantum mechanic confinement significantly increases thermopower. The complex crystal structure of clathrates based on polyhedral cages intrinsically provides the desired length scales in the nanometer range, comparable to mean free paths of typical phonons. Further reduction of lattice thermal conductivity is brought about by the introduction of larger length scales via nanotechnology such as melt spinning, ball milling, or substitutions to create defects at nanometer distances. This ensures that the heat carrying phonons will be scattered by objects at a variety of proper length scales. For optimal figure of merit, renormalization of details of the electronic density of states at the Fermi energy (E_F) will be achieved via specific resonance states near E_F. Introducing virtual bond states as provided by 3d, 4d, 5d elements (e.g. Mn, Fe, Co, Ni, Pd, Pt etc) might closely follow this concept (like Tl in Bi_2Te_3, Heremans, Science 321, 554 (2008)).

Aspects of nanostructuring will cover formation of layered structures by MBE, sputtering, or thermally evaporated thin films with superlattices. This will infer direct interaction of experimental work with theoretical modelling on a microscopic level provided by band structure calculations (LDA and tight binding approach, phonon dynamics and phonon interaction) .

High efficiency thermoelectrics

The envisaged research aims towards the design of a novel dense high-efficiency thermoelectric generator (TEG) on the basis of low cost materials like type I Si-based clathrates. As thermoelectric performance basically depends on two factors, (i) the temperature gradient used (Carnot –efficiency) and (ii) the thermoelectric efficiency ZT, the high melting temperature of Si-based clathrates is a perfect prerequisite of hot stage operation temperatures even above 600°C.

So far V. Zlativ (Zagreb), E. Bauer, S. Bühler, Pachen,K. Held (all TU Wien), E. Rogl (Universität Wien) and N. Böttner (Freiburg) have expressed their interest to join the project, but we aim at including further European scientists.